



PATENT APPLICATION

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Applicants: Hironori KOBAYASHI et al

For: RESIN COMPOSITION

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LETTER TO EXAMINER

Sir:

At the outset, Applicants' representative wishes to thank Examiner Dollinger for the courtesy exhibited during an interview conducted on June 9, 2010. During the interview, it was discussed whether a showing of unexpected results was necessary to overcome the prior art cited against the present claims and whether this type of evidence was already of record in the present application. Examiner Dollinger indicated that he would consider any unexpected results already of record or submitted in the present application but the unexpected results must be commensurate with the scope of the claims. Applicants' representative indicated that he would attempt to present such a showing during the further prosecution of the present application.

Claims 1-3, 5-9 and 11-13 have been rejected under 35 USC 103(a) as being unpatentable over Takei et al in view of

Kumagai. Applicants respectfully traverse this ground of rejection and once again urge that the presently claimed invention clearly is patentably distinguishable over these references.

As stated previously, the instant invention is directed to a resin composition comprising a polyol, a polyisocyanate and a silane coupling agent containing an imidazole group as essential components. The ratio NCO/OH of the number of isocyanate groups in the polyisocyanate (B) to the number of hydroxyl groups in the polyol (A) and the composition is from 0.6 to 4.0 and the weight ratio of [(A)+(B)]:(C) is from 100:0.01 to 100:10 and the silane coupling agent containing an imidazole group (C) is the reaction product of a reaction mixture consisting of either (1) an imidazole compound and 3-glycidoxypolytrimethoxysilane or (2) an imidazole compound and 3-methacryloxypropyltrimethoxysilane and has a hydroxyl group in its molecule when it is obtained by reaction of an imidazole compound and 3-glycidoxypolytrimethoxysilane and does not have a hydroxyl group in its molecule when it is obtained by a reaction of an imidazole compound and 3-methacryloxypropyltrimethoxysilane.

In the present invention, the silane coupling agent containing an imidazole group reacts with a polyisocyanate or a polyol and forms a three-dimensional network in the resin to enhance the properties of the cured resin. Of silane coupling agents having an imidazole group, those having a hydroxyl group in their molecular are advantageous in that the portion with the hydroxyl group reacts extremely efficiently with a polyisocyanate to create a more developed network structure so that the compounds are particularly good in terms of improving the adhesion to base materials. The present inventors have found, for the first time, the fact that the silane coupling agent containing an imidazole group is itself involved in the formation of a three-dimensional network in the resin so that a more developed network structure can be formed.

In contrast thereto, nitrogen atoms in the imidazole group of the silane coupling agent containing an imidazole group simply have the effect of promoting the curing reaction of an epoxy resin, just as tertiary amine compounds have for epoxy resin curing, and silane coupling agents containing an imidazole group just remain in the resin without participating in the resin's network formation by polymerization. In the present invention, alkoxy silyl groups or hydroxyl groups contained in the silane coupling agent containing the imidazole group react with hydroxyl groups in a polyol and isocyanate groups in polyisocyanates, respectively, to form a three-dimensional network by polymerization in the urethane formation as well as a polymerization mediated by the silane coupling agent. Once again, it is respectfully submitted that the prior art cited by the Examiner does not disclose the presently claimed invention.

The Takei et al reference discloses a process for producing a polymer having a hydroxyl group at both terminals by polymerizing a vinyl-based monomer in the presence of an alcohol having no addition-polymerization reactivity through the use of an initiator consisting essentially of a peroxide and a catalyst selected from the group consisting of an organic sulfonic acid compound having no addition-polymerization reactivity and an inorganic acid. Specifically speaking, this reference discloses the polymerization of a vinyl-based monomer in the presence of an alcohol having no addition-polymerization reactivity through the use of the initiator consisting essentially of a peroxide and at least one catalyst selected from the group consisting of an organic sulfonic acid compound having no addition-polymerization reactivity and an inorganic acid. Prior to the reaction, there is no disclosure in this reference regarding the mixing of a silane coupling agent containing an imidazole group with at least one of the polyol and the polyisocyanate. In fact, this reference specifically discloses that the reactor does

not substantially contain any other component than the named vinyl-based monomer, alcohol, peroxide and catalyst.

The Kumagai et al reference is directed to an imidazole/organic monocarboxylic acid salt derivative reaction product which improves the adhesion between a resin and a metal such as copper, steel or aluminum, or an inorganic material such as glass fiber, silica, aluminum oxide or aluminum hydroxide. Example 3 of this reference discloses 100 parts of an epoxy resin, 5 parts of dicyandiamide, 2-ethyl-4-methylimidazole and one part by weight for bonding together two surface-treated aluminum alloy plates through the formation of a resin composition having an improved mechanical strength.

Column 5 of this reference discloses that the imidazole/organic monocarboxylic acid salt derivative reaction product can be used as a resin additive or a surface treatment agent for improving adhesion and polyurethane is specifically named as such a resin. However, there is no suggestion in this reference of adding the silane coupling agent containing an imidazole group to a polyol and/or a polyisocyanate prior to their reaction to form the polyurethane resin.

The nitrogen atoms in the imidazole group of the silane coupling agent of Kumagai et al simply have the effect of promoting the curing reaction of the epoxy resin, just the same as tertiary amine compounds due in epoxy resin curing, and the silane coupling agent containing the imidazole group remain in the resin without participating in the resin's network formation by polymerization. This reference merely shows that the above-mentioned effect most effectively takes place during epoxy resin curing. In Kumagai et al, the epoxy resin and the curing promoter react sequentially, making the resulting polymer substance have a two-dimensional structure as opposed to a three-dimensional structure as is formed by the present invention. Claims 11 and 12 are even further distinguished over the prior art cited by the Examiner as Claim 11 consists essentially of the polyol, the

polyisocyanate and the silane coupling agent and Claim 12 consists of the polyol, the polyisocyanate and the silane coupling agent. The references cited by the Examiner require that other critical ingredients be present. Therefore, Claims 11 and 12 are even further patentably distinguishable over the prior art cited by the Examiner.

Although the Examiner has not made a showing of prima facie obviousness under 35 USC 103(a) with respect to the presently claimed invention, objective test data is of record in the present application which is more than sufficient to overcome any proper rejection under 35 USC 103(a).

On pages 8-14 and in Figure 1 of the present specification, objective test data is presented which shows the unexpected benefits gained by using the silane coupling agent containing an imidazole group of the present invention. In Examples 1 and 2 and Comparative Examples 1-4, the curing promotion effect of an imidazole group-containing silane coupling agent of the present invention was evaluated against 3-glycidoxypolytrimethoxysilane as a common silane coupling agent in Comparative Example 1, 2-ethyl-4-methylimidazole as a conventional imidazole catalyst in Comparative Example 2 and triethylamine in Comparative Example 3. As discussed in paragraph [0029], in the reaction system in which an imidazole group-containing silane coupling agent was added, the residual isocyanate group content was lower than that of the system to which no catalyst was added.

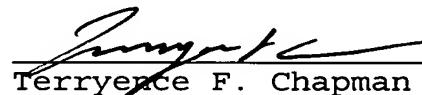
In Examples 3 and 4 and Comparative Examples 5-11, the contribution of the imidazole group-containing silane coupling agent to the increased adhesion to a base material was evaluated. Examples 3 and 4 use imidazole silane compounds according to the present invention while Comparative Example 5 used 3-glycidoxypolytrimethoxysilane, Comparative Example 6 used 3-aminopropyltriethoxysilane, Comparative Example 7 used 3-mercaptopropyltrimethoxysilane, Example 8 used 2-ethyl-4-methylimidazole and Comparative Examples 9 and 10 used tertiary amines. As shown in the results contained in

Table 1, the imidazole silane compounds of the present invention resulted in the resin composition exhibiting superior adhesion. This is clearly not suggested by the prior art cited by the Examiner.

The corrosion resistance effect of the imidazole group-containing silane coupling agent of the present invention was evaluated in Examples 5 and 6 and Comparative Examples 12-15. In Examples 5 and 6, imidazole silane compounds of the present invention were used while Comparative Example 12 used 3-aminopropyltriethoxysilane, Comparative Example 13 used 2-ethyl-4-methylimidazole and Comparative Example 14 used a tertiary amine. As shown in Table 2, the imidazole silane coupling agents of the present invention resulted in an increased corrosion resistance as compared to the comparative additives while still eliminating the unpleasant odor associated with conventional tertiary amine catalysts. The test results discussed above are clearly sufficient to establish the patentability of the presently claimed invention over the prior art cited by the Examiner.

The Examiner is respectfully requested to reconsider the present application and to pass it to issue.

Respectfully submitted,

  
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Encl: None

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